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The Electrification of Ice and Supercooled Water Drops

by

D.G. Evans, B.Sc.

Presented in candidature for the degree of Master of Science

July, 1962

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ABSTRACT

The electrical structure of a thunderstorm consists essentially of a positive dipole, with values of the segregated charge estimated at 20 coulombs. Between the main charge centres there exists mixed charge, of both signs, of the order of 1000 to 1500 coulombs. As this charge is generated in regions where the temperature is well below $0^{o}C$ the phenomena have been associated with the charging of ice.

In recent years several workers have found that if a temperature gradient is set up in ice, the colder part acquires a positive charge, and the warmer a negative charge. This has been explained by assuming that protons migrate from the warmer to the colder part of the ice, and Mason and Latham (1961 a) have put forward a quantitative theory, which is in very good agreement with their experimental results.

In the present work the charging due to the momentary contact of vapour grown ice crystals at different temperatures was investigated. Ice crystals were grown in a diffusion chamber on a fine insulating fibre and a fine horizontal wire supported by a cam driven rod. Contact could be made between ice crystals grown at different levels in the diffusion chamber and the fibre then raised into a bronze Faraday cylinder, which was connected to a Vibron electrometer

adapted for measuring single charges. Contact times of the order of $\frac{1}{100}$th second were employed, this being the time for maximum charge separation predicted by Mason and Latham's theory. However, although temperature differences of up to $10^{\circ}C$ were involved no charging was observed.

When water droplets are nucleated just below $0^{\circ}C$ a strong ice shell is formed and further freezing produces a pressure inside the drop which finally causes the drop to burst. Mason and Maybank (1960) measured the charging which accompanies the bursting and attributed it to the migration of protons along the radial temperature gradient set up in the ice shell; the charge separation being caused by an excess of either the outer or inner surface of the shell carried away when the drop bursts. Mason and Latham (1961) suggest that this process may account for the charging of a hail pellet growing by the accretion of supercooled droplets.

An attempt to measure the charging of a single supercooled droplet, brought into contact with an ice surface, was unsuccessful, but the charging of nucleated supercooled droplets was observed. One millimetre diameter droplets suspended from the insulating fibre were cooled to just below $0^{\circ}C$ and then nucleated by dropping a small piece of solid carbon dioxide into the diffusion chamber. The droplets were raised into the Faraday cylinder after freezing was completed and no charging was observed unless fragmentation of the drop had taken place.

The results obtained suggested that the charging was too large to be accounted for by the temperature gradient theory, and also that the sign of the charge remaining on the drop was determined by the amount of water present when the drop bursts.

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CHAPTER ONE

METEOROLOGICAL AND ELECTRICAL PHENOMENA IN THUNDERSTORMS

A thunderstorm is characterised by extreme turbulence and intense electrical disturbances. It is intended to give here a brief account of the meteorological and electrical phenomena, which are present in thunderstorms, so that the theories to be discussed in the next chapter may be fully appreciated.

1. Meteorological Phenomena

A thunderstorm consists of one or more distinct cells (average diameter 1 Km.) with intense air motions, precipitation and electrical activity, which are separated by relatively still air. The air motions are localised and "sub cells" (300 m. in diameter) have been described, which account for the turrets seen to rise from the top of a single cell.

The life of a cell is divided into three stages determined by the direction and velocity of the air motions. The developing or cumulus stage has up draughts extending from ground level to the cloud top, with average velocities of 5 m./sec. and maximum velocities of 15 m./sec. Dry air is drawn into the cell at all levels due to pressure differences and exchanges of momentum between the rising air and the environment. This 'entrainment' of air occurs in

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all three stages and in the cumulus stage tends to decrease the accelerating force as cloud droplets evaporate and the temperature is lowered. The hydrometeors present in this stage are small, and liquid water exists for a few thousand metres above the $0^{\circ}C$ isotherm. Higher up there is mixed rain and snow, wet snow and finally dry snow. The electric field, measured at the ground, develops at the time of the first radar return and increases exponentially until the first discharge. During this stage the visible cloud top reaches a height of 10 km. at a temperature of $-40^{\circ}C$ with the cloud base at approximately $+10^{\circ}C$. This stage lasts for 10 to 15 minutes after the first radar echo.

The hydrometeors grow by condensation and finally reach a mass which can no longer be supported by the up draught. The drag produced by the fall of the hydrometeors causes the development of downdraughts in the lower and middle regions of the cell, which is now in the mature stage. The first precipitation at the cloud base coincides with the change from the cumulus to the mature stage. (It is during this stage that the most intense effects at the ground occur, heavy rain and strong gusty winds being common). The down draughts increase in force both vertically and horizontally and begin to spread throughout the cloud. Rain is found in the lower levels, mixed rain and snow above and finally dry snow. The visible cloud top reaches its maximum in this stage usually between 10 and 15 km.

As the radar top begins to move downwards intra-cloud lightning discharges take place increasing in intensity and frequency until after about 7 minutes cloud to ground flashes occur. The whole stage lasts from 15 to 30 minutes.

The down draughts continue to spread throughout the cell at higher and higher levels until the whole of the cell contains only down draughts or no vertical motion. This, the dissipating stage, lasts a further 30 minutes and after it the storm has ended.

Recently Ludlam (1961) has made a detailed study of a frontal storm over South East England and the analysis gives results which differ from those described above. The observations indicate that the up draughts and down draughts act continuously side by side and that each is necessary to maintain the other. The up draught is fed by warm, moist air entering the front of the thunderstorm and the down draught by cold, dry high level air entering from behind. Thus, the up draught current rides over the down draught, the two being separated by a sloping surface. Rain or hail falling from the up draught will tend to evaporate in the dry environment of the down draught thus causing the air to be chilled and accelerating the down draught. In this particular storm the lifting of the warm air was carried out ahead of the storm by the cold air spreading out from the rain area. In this way the storm once established could maintain itself. The growth of hydrometeors is also discussed and a mechanism is described by which a hailstone can be recirculated until it reaches a size large enough to fall out of the up draught.

2. Electrical Structure

Measurements have been made of the potential gradients produced by thunderclouds both at ground level and inside the cloud. As the potential gradients are produced by the free charges inside the cloud values can be obtained of both the magnitude and position of these charges. The results show that a thundercloud possesses essentially a vertical positive dipole with in many cases a smaller, lower positive charge. It is thought that this lower positive charge, which is associated with heavy rain may, in fact, exist in all thunderstorms.

The upper positive charge occupies a diffuse volume at approximately $-20^{\circ}C$ and may be displaced from the axis of the main dipole by wind shear. The negative charge is much more concentrated and is situated around the $-5^{\circ}C$ isotherm. It lies in the same vertical line as the lower positive charge, which occurs at, or just below, the $0^{\circ}C$ isotherm. It is of interest that the negative charge is situated in the precipitation zone, but does not extend down into the rain sheet.

Values of the charges present have been deduced from potential gradient measurements on the ground and these indicate that the main dipole consists of 20 - 50 coulombs separated by 2 - 5 kms. The lower positive charge is some 2 kms. below and has a value of about 10 coulombs. It should be mentioned that these are values of charges that have been separated and that there exist large quantities of charges of both signs between the two centres.

Most of the theories for the production of the main dipole consider that gravitation is the segregating process. This means that positive and negative charges are first attached, in some way, to different carriers, which then move relatively to one another under the influence of gravity. The negative charges may be carried on heavy precipitation particles while the positive charges move upwards in the up draught on much lighter particles. The gravitational segregation continues until the field has built up to the value required for discharge. After a lightning flash the field builds up exponentially reaching very nearly its predischarge value in 7 seconds. To account for this rate of charging it has been calculated that the quantity of unseparated charge between the two centres must be between 1,000 and 1,500 coulombs.

Vonnegut and Moore (1958) have put forward a theory for the charge segregation that does not involve gravitation. It is suggested that atmospheric space charge is drawn up into the base of the cloud by the convection currents, and that the small electric field set up causes a conduction current to flow in the clear air, which produces a layer of opposite charge around the cloud. This is then carried in the down draughts so that a continuous current is set up between the upper atmosphere and the lower part of the cloud. As the field builds up point discharge occurs and the process is strengthened.

The theories discussed in the following chapters all consider the gravitational process and involve the ice phase in the mechanism of both generation and segregation of charge.

CHAPTER TWO

GENERATION OF CHARGE IN THE ICE PHASE

Results from the altielectrograph measurements made by Simpson and Serase (1937) showed the existence of the upper positive charge centre in regions where the temperature was well below 0°C. Earlier Simpson had measured large potential gradients, usually positive, which were produced by blizzards in the Antarctic and his explanation of these was applied to a charge generation in thunderstorms. Simpson (1942) explains the mechanism of the theory; turbulence in the thundercloud causes the frequent collision of ice crystals, which gain a negative charge, an equal positive charge going as ions in the air. The ions then become attached to cloud particles and the charges are separated by gravitational forces. Simpson believed that the charging was due to tiny ice splinters being broken from the delicate structure of the ice crystals during impact.

1. Charging due to impact and friction

Since Simpson and S~~e~~crase put forward their theory, experimental work done on the charging of ice due to frictional contact has often been contradictory. Pearce and Currie (1949) eroded a block of snow with an air blast and found that the charging was small until the surface of the block was fractured. Then considerable charge was produced, the eroded fragments being negatively charged and the air receiving a positive charge. Stimmel et al (1946) showed that when snow is blown against various targets large amounts of charge are produced, but the nature of the target determines the sign of the charge. Norinder and Siksna (1954) carried out similar experiments and came to the same conclusions. However, when a snow block was used as the target the results were opposite to those of Pearce and Currie.

Impact velocities expected in the atmosphere were employed by Chapman (1949) who measured the charging produced by a single snowflake as it fell into a dish of snow. The measurements were made in actual storms and the collector had a collimating device to separate single flakes. Measurements were made of the initial charge on the snowflake, the charge acquired by the dish after the snowflake had fallen on it and the charges in the air. Apparently the magnitude of the charges produced varied widely, but one result, which was reported to be typical, showed that the dish acquired a positive charge of 10^{-12}c, and negative charge was observed in the air. If only the magnitude varied widely, Chapman's results give the wrong sign for thunderstorm electrification. Chalmers (1952) carried out an experiment, which showed the correct separation. Two handfuls of snow were rubbed together and the fragments were allowed to fall into a collector which was connected to an electrometer. A negative charge was always indicated, the positive ions presumably being removed by the wind. Separate experiments ruled out the possibility that charging by induction, or contact with the metal collector was responsible for more than a small charge and friction appeared to be essential for the large charges observed.

2. <u>Experiments involving temperature gradients</u>

Workman and Reynolds (1954) carried out the first of a series of experiments, which have led to a quantitative theory, which can be tested experimentally and may account for the contradictions in the earlier work. The electrical effects accompanying the growth of precipitation particles were investigated and the results indicated that the collision of ice crystals with the growing ice particle was of prime importance. The apparatus consisted essentially of two ice covered metal spheres rotating with the velocity of a 4 mm. diameter graupel particle (about 8 m/sec.) in a supercooled cloud. The composition of the supercooled cloud could be varied so that it consisted of supercooled droplets, supercooled droplets and ice crystals in coexistence and ice crystals alone. The highly insulated spheres were connected via a mercury contact to an electrometer probe. When the cloud consisted of supercooled droplets or ice crystals alone negligible charging was observed, but when the two occurred together large amounts of charge were separated, the sign depending upon the relative concentrations. Positive charging of the spheres occurred when there was a predominance of ice crystals and negative charging when supercooled droplets were in excess.

Under conditions for negative charging the ice on the spheres will be growing by accretion of droplets and will thus be warmer than the environment because of the latent heat liberated. To test whether this temperature difference was of importance conditions were set for positive charging, but the spheres were heated by a lamp. Strong negative charging resulted immediately. The sign of the charging was thus shown to be strictly controlled by the temperature difference. Henry (1953) had put forward this idea earlier to explain the separation of charge when two insulating materials were rubbed together. He suggested that the temperature gradient, produced by asymmetric rubbing of the two surfaces, results in the more mobile particles moving from the hot to the cold side of the boundary, the phenomenon being analogous to the thermal diffusion of gases and the Thomson thermoelectric effect.

Workman and Reynolds (1954) set up an experiment to test this hypothesis. Films of ice were formed on two nickel plated copper rods, one of which was earthed and the other connected to an electrometer. They found that when the two were rubbed together the warmer always acquired a negative charge. Further experiments were carried out with impurities in the ice films and here it was found that the ice with the greater proton mobility (i.e. the higher conductivity) acquired a negative charge even when several degrees colder than the other ice film. Workman et al (1954) have found that conduction in ice depends upon a proton transfer mechanism and these results indicate that proton migration along the temperature gradient is the cause of the observed charge separations.

The experiments of Workman and Reynolds involved frictional contact of the ice surfaces, but Brook (1958) carried out work in which rubbing and frictional contact were minimised. One rod was suspended from the probe of an electrometer and maintained at a constant temperature inside a large aluminium block, which was fitted with a heat sink and heater. The second rod was supported on a motor driven carriage and contact was made by moving the rod in and out of a hole in the aluminium block. The temperature of the second rod was varied by placing heated blocks on the rod and allowing them to ride with the carriage. A continuous record of the 'probe ice' potential was made during the experiments. The results showed that the potential difference between the two pieces of ice while in contact was sensitive to the temperature difference.

between them. In one experiment the probe ice was maintained at $-24^{\circ}C$ while the temperature of the second rod changed from $-27^{\circ}C$ to $-21^{\circ}C$; the record shows a reversal of potential immediately the temperature difference changed from positive to negative. The experiments show that a potential difference can be produced by a temperature gradient alone, frictional contact not being necessary.

The mechanism put forward to explain the experimental results just described can be summarised as follows; if two pieces of ice with different conductivities are brought into contact protons will move to equalise the conductivities. The difference in conductivity may be due to either temperature or contamination differences. Thus if the contact is momentary a charge separation will occur, the ice with the greater conductivity losing protons and being left with a resultant negative charge.

A qualitative theory for the charge generation in thunderstorms has been put forward by Reynolds et al (1957) on the strength of these results. A graupel particle growing by accretion of supercooled droplets will be warmer than crystals growing by sublimation due to the latent heat liberated. Also the droplets will be contaminated by their condensation nuclei and so the graupel particle will consist of ice with a greater conductivity than the purer ice crystals. Therefore, both the variables are in favour of the graupel becoming negatively charged after collision with an ice crystal. It is suggested that gravitational segregation then occurs with the graupel particle carrying the negative charge and the lighter ice crystals the positive charge.

3. Mason's work

Latham and Mason (1961 a) have recently put forward a quantitative theory of the proton transfer mechanism and have tested the theory experimentally. In ice the proton plays a part similar to that of the electron in metallic conduction and the theory is based on two known facts (1) the concentration of H^+ and OH^- ions in ice rises rapidly with increasing temperature, (2) the mobility of the H^+ ion is at least an order of magnitude greater than that of the OH^- ion.

The first case considered is that of a steady temperature gradient maintained across a layer of ice. The charge separation is described in the following way: The (temperature) gradient will lead to a separation of charge with a net excess of positive charge in the colder part of the ice. The space charge set up by this differential diffusion will produce an internal electric field, and a steady state will be reached in which no net current flows, the diffusive flux of ions maintained by the temperature gradient being balanced by a reverse current maintained by the potential gradient. Equations are derived for the currents due to the positive and negative ions and assuming that the internal electric field is uniform an expression is obtained for the charge separated. The expression evaluated gives $q = 4{\cdot}95 \times 10^{-5} (\frac{dt}{dx})$ e.s.u. per cm.^{2}, where 'q' is the charge separated and $\frac{dt}{dx}$ the temperature gradient in the ice.

The second case considered is that in which two pieces of ice at different temperatures are in momentary contact. Here it is shown that the charge separation is dependent upon the time of contact and that the maximum charge is obtained for the time of contact of approximately 10^{-2} seconds. The maximum charge $q_{max} = 3{\cdot}05 \times 10^{-3}(T_1 - T_2)$ e.s.u. per cm.^{2} where q_{max} is the charge separated per unit area and T_1 and T_2 the temperatures of the two ice surfaces. It is shown that as the time of contact is prolonged the charge separated diminishes as the temperature gradient is destroyed, and ultimately the pieces of ice become neutral.

In order to measure the charging due to steady temperature gradients a specimen of ice was frozen onto two copper discs and these were placed between, but insulated from, two copper rods. The rods could be heated or cooled separately and were used to provide the temperature gradient across the ice. The copper discs act as a condenser with the ice as the dielectric and the charges separated were calculated by measuring the potential differences developed between the discs, one being connected to earth and one to an electrometer. From the results a graph was plotted of potential developed against temperature difference. A linear relationship was obtained the values being in good agreement with the equations derived from the theory. The influence of contaminants was investigated and it was found that whereas small amounts of HF and CO_2 increased the charge separation, Na Cl caused a decrease. This decrease has been explained by Mason as being due to concentrations of salt at the crystal boundaries setting up leakage paths in the ice.

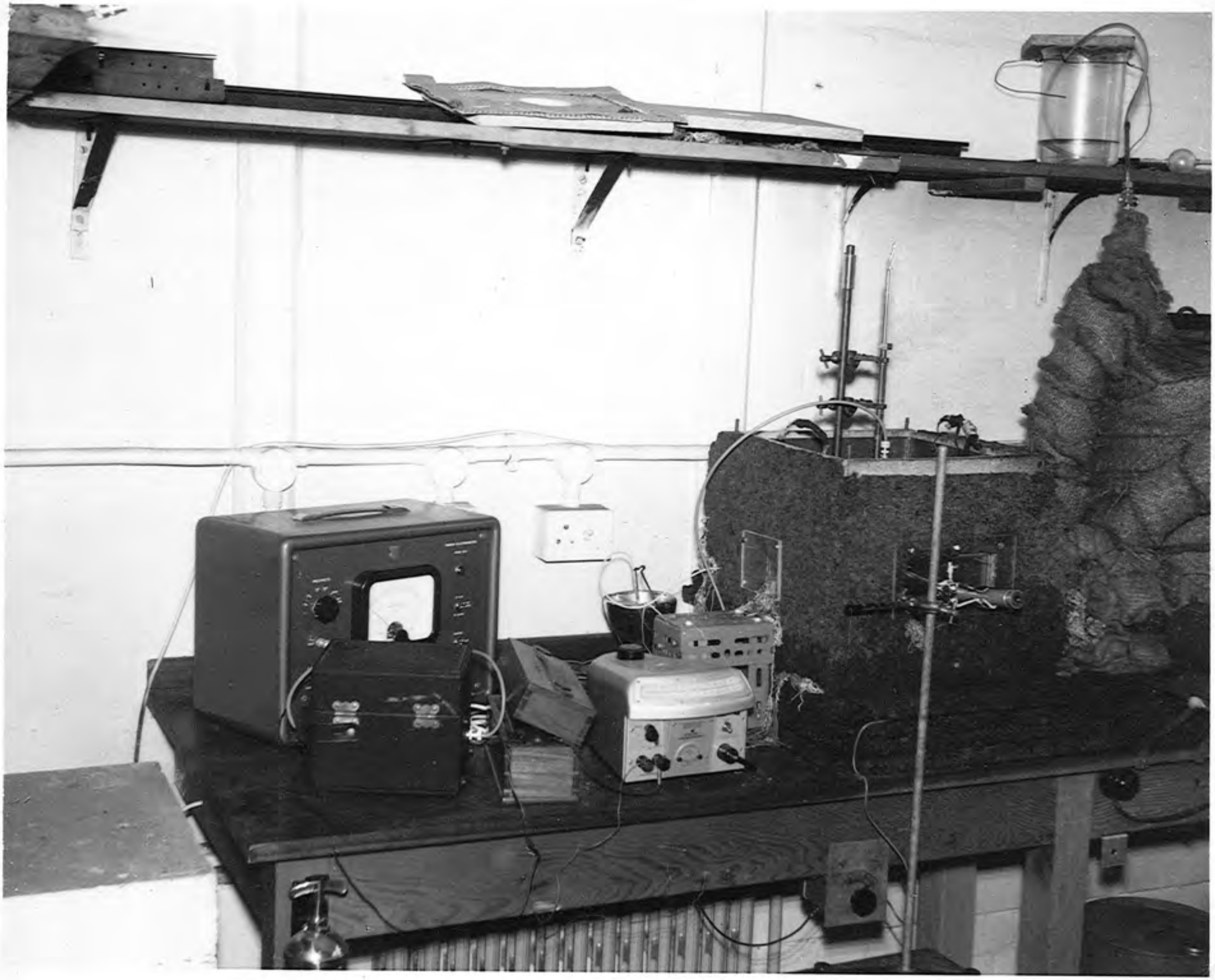
Bouncing contact for 10^{-2} seconds was achieved by using an earthed cam driven rod, which was brought into contact with a similar rod connected to an electrometer. The 'probe ice' was held at room temperature ($-16^{\circ}C$) and the temperature of the moving rod could be varied from 0 to $-40^{\circ}C$. The capacitance of the system was calibrated so that measurement of the potential developed during contact allowed the charge separated to be calculated. The results were in good agreement with the values of the charge separation predicted by the theory. By varying the time of contact it was shown that for times greater than 10^{-2} seconds a decrease in the charging occurred and at 0·5 seconds no charge separation was detected. The introduction of contaminants (HF.,NaCl, and Co_2) into the warmer ice increased the charge separation, but decreased it when placed in the colder ice.

The very good agreement between the experimental results and the values predicted by the theory gives strong support to the proton transfer mechanism. The impurities give greater charge separation when placed in the warmer ice as they increase the concentration gradient. However, when placed in the colder ice the impurities decrease the concentration gradient and can in fact reverse it (Brook 1958).

Latham and Mason (1961 b) went on to investigate the charging found during experiments with growing rime ice. Two separate experiments were carried out, one on the charging due to ice crystal collisions and one on the charging due to impinging water droplets.

In order to eliminate the effects due to water droplets the 'hail pellet' consisted of a 0.5 mm. coating of ice previously grown on an insulated metal cylinder and the ice crystals were produced by passing air saturated with water vapour over solid carbon dioxide. The temperature of the ice probe was varied by means of a coolant vessel and heater and was measured by means of a thermocouple. The air and ice crystals were drawn past the ice probe and their temperature was measured just before reaching the ice probe. An induction cylinder connected to an electrometer was placed below the ice probe, which could be lowered into the cylinder at intervals to measure any charge accumulated. It was found that the probe always acquired a negative charge if it was warmer than the ice crystals and a positive charge if colder. The presence of impurities gave exactly similar results to those in the bouncing contact experiments. An estimate showed that a crystal of 20 μ diameter, which was $5^{o}C$ colder than the ice probe gave a separation of $- 5 \times 10^{-9}$ e.s.u. per collision.

Using the value of $- 5 \times 10^{-9}$ e.s.u. per collision Mason calculated that charging in a thunderstorm due to ice crystal contact alone would give a value four orders of magnitude lower than the observed charging. His results on the charging due to the accretion of water droplets, give the correct order of magnitude when applied to thunderstorms and this work will be described in a later chapter.



The Apparatus

CHAPTER THREE

THE APPARATUS

The apparatus was designed initially by Hutchinson (1960) to measure the charging produced by single ice crystal contacts. It consists essentially of a cooling system, which produces a vertical temperature gradient inside a diffusion chamber and an arrangement for measuring single charges. The three main parts, their performance and operation will be described in this chapter.

1. The cooling system

The large copper base of the diffusion chamber is cooled by continuously circulating paraffin oil, previously cooled in the tank A, the whole arrangement being shown in Figure 1. The base was designed so that a surface of uniform temperature was presented to the diffusion chamber. This was achieved using a tank 12" square and 4" deep, with a top of $\frac{1}{8}$" copper sheet, permanently filled with paraffin oil. The paraffin oil is circulated through the base by two copper tubes, which run close to opposite walls and have holes facing outwards towards the walls.

The circulating paraffin is cooled by passing it through a fairly close helix of copper tubing inside the large tank A in Figure 1. The tank, 20" high and 10" in diameter, has fixed inside it a metal frame 8" in diameter around which the helix is wound. The paraffin oil in the tank is cooled by a

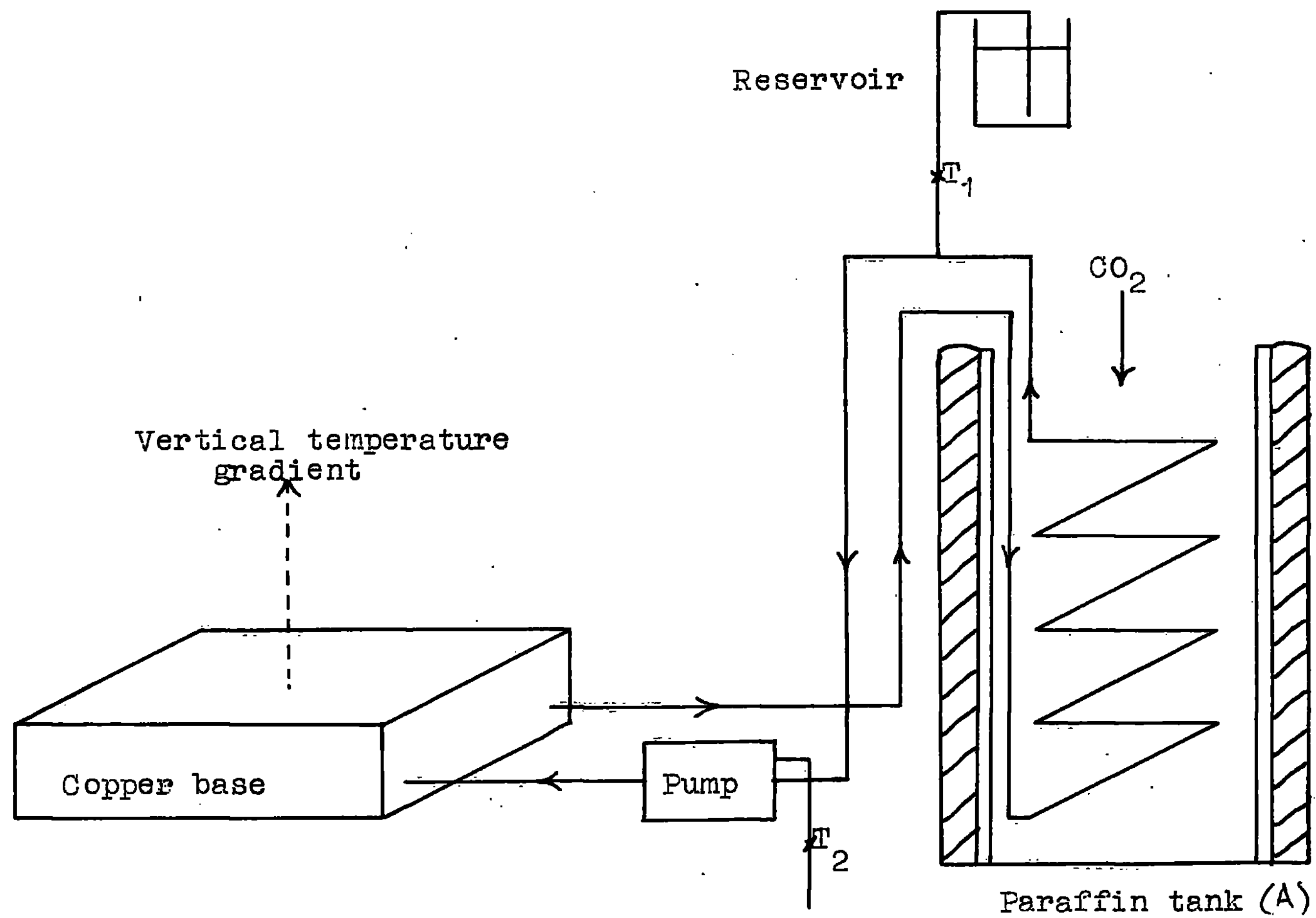


Fig. 1 The Cooling System

full block of solid carbon dioxide, which fits inside the metal frame and produces a temperature of around $-40^{o}C$. It was found that higher fractions in the commercial paraffin oil solidified around the block, forming a thick insulating layer, which had to be scraped away to prevent the tank from warming up. It is important, therefore, that the solid carbon dioxide does not come into contact with the copper helix in order to avoid a blockage in the circulating system.

Before the block of solid carbon dioxide is put into the tank, it is necessary to cool down the paraffin oil with a few small pieces of solid carbon dioxide as the gas liberated causes the paraffin oil to bubble vigorously and to rise rapidly in the tank.

Figure 1 shows the closed system through which the cooled paraffin oil is circulated by the centrifugal pump. A reservoir is provided so that as the paraffin oil contracts on cooling more can run in to keep the system full, tap T_1 being always kept open. This is necessary to prevent air blockages, which could stop the flow of paraffin oil. (An air blockage did in fact occur due to a leak in the pump and the cold paraffin oil was pumped up into the reservoir). The problem of blockages is a real one and when filling the closed system of tubes and base care must be taken that all the air is removed and that no dirt is allowed to enter.

The procedure for filling the system is as follows. A rubber tube is attached below tap T_2 and immersed in a large beaker of carefully filtered paraffin oil. A thistle funnel is fixed to the tube above T_1 and paraffin oil sucked up into it. Tap T_2 is then closed and the thistle funnel is removed with the tube below the level of the paraffin oil in the reservoir. In this way paraffin oil is siphoned from the reservoir into the tubes and copper base. A plug in the top corner of the copper base is removed and this corner raised so that the plug hole is at the highest point in the base. Air is thus forced out of the base until it is completely filled with oil. The plug is then replaced and the centrifugal pump switched on. This drives any air bubbles present up into the reservoir. The last two steps are repeated until no air bubbles are seen to leave the system.

The whole system stands on four layers of 1" cork sheets, which gives good insulation and also reduces vibrations from the pump. The paraffin tank and copper tubes are tightly wrapped in hessian backed felt insulation and a chip-board cover is fitted to the top of the tank.

The cooling system was fairly satisfactory apart from the problems of leaks and blockages. A temperature of around $-30^{\circ}C$ was reached by the base in 2 to 3 hours after the introduction of the solid carbon dioxide into the tank, and the temperature gradient in the diffusion chamber remained

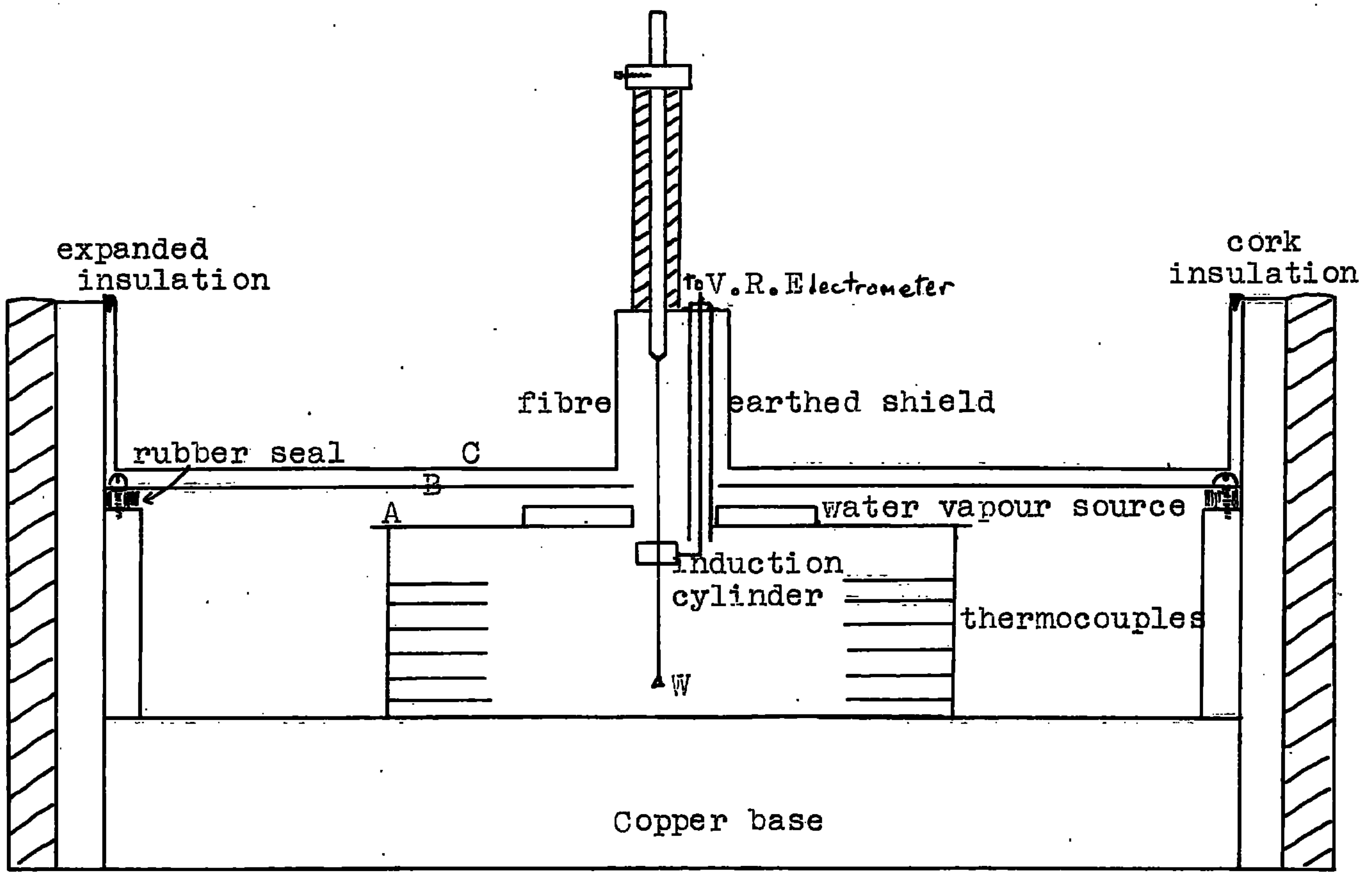


Fig. 2   Diffusion chamber

constant throughout the day. The main disadvantage of this system is that the temperature range is limited to 0 to $-30^{\circ}C$ although this did not really affect the present work.

2. <u>The diffusion chamber</u>

The complete diffusion chamber is shown in Fig. 2. The outer walls are made of $\frac{3}{4}$" chip-board covered with a 1" layer of cork, which gave adequate insulation. The overall dimensions of the chamber are 12" x 12" x 13". The back and front walls have perspex windows for illumination and viewing respectively, with double air spacing to minimise heat flow. Illumination is provided by a 60 watt table lamp placed behind a glass tank filled with water to absorb the heat. A microscope with a 3" objective was constructed for viewing and measurements inside the chamber. Later a small perspex window, also with double air spacing, was built into a side wall to allow side illumination to be used for photographic purposes.

The inner compartment, in which the experiments were carried out, is made of perspex and was built in the centre of the main chamber to minimise convection currents. The construction of this chamber is shown in Fig. 3. The sides A and B run from the front to the back of the main chamber and are firmly fixed to the outer walls. The other sides were tapped and bolted on to A and B forming an inner chamber

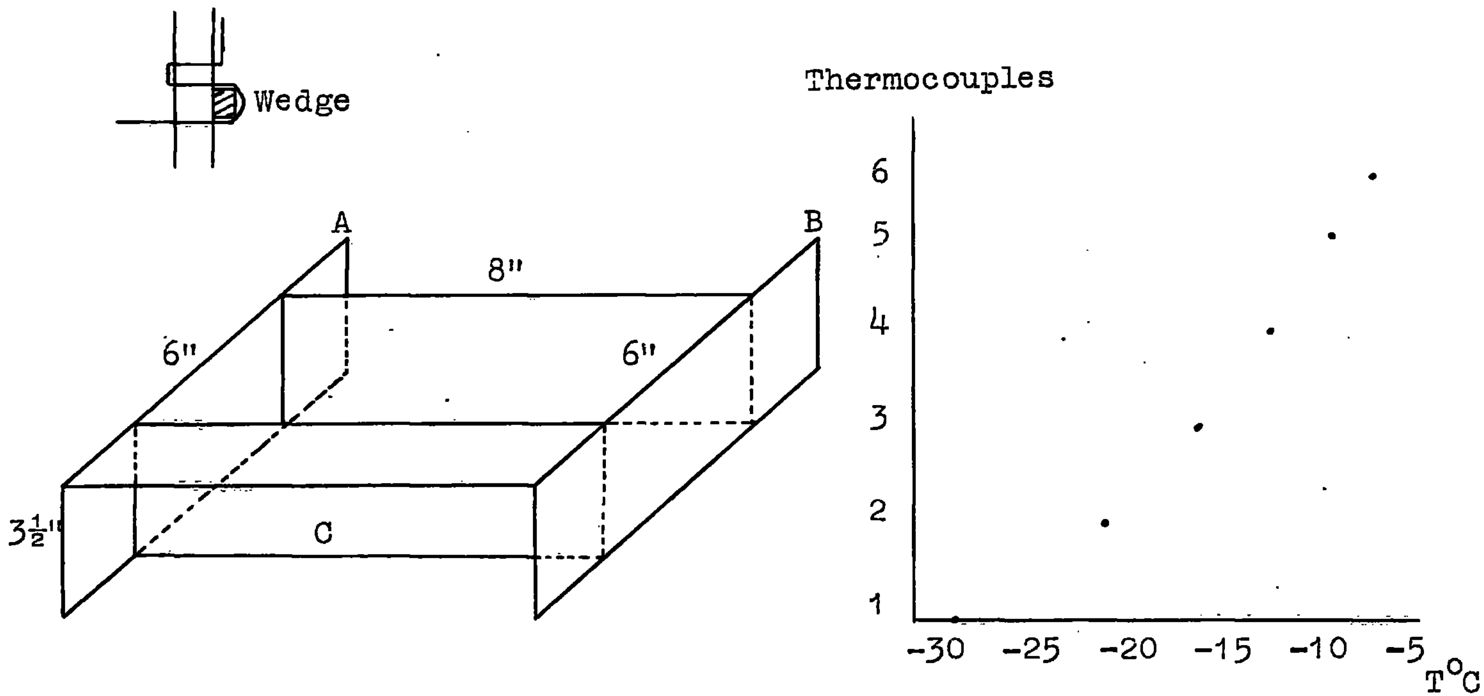


Fig. 3 The Inner Chamber

6" x 8" x $3\frac{1}{2}$". All the joints and contacts with the copper base were sealed with Durofix adhesive. This was done to prevent the influx of water vapour which had previously condensed on the inner walls of compartment C near the viewing window.

In order to measure the temperature gradient six copper-Constanton thermocouples of 30 S.W.G. wire were arranged horizontally with one junction at the centre of the chamber. To prevent the wires from sagging when the chamber was cooled down the wires were held taut by small wedges placed between the wires and the chamber wall. The position of the wedges and the way in which the wires were secured is shown in the enlargement in Fig. 3. The complete thermocouple arrangement is shown in Fig. 4. Each thermocouple has one junction in the chamber and one maintained at room temperature inside a brass block, which stands in a Dewar flask filled with water. The thermocouples are connected across a variable resistance (R in Fig 4) and a Scalamp galvanometer in series. The selector switch S allows each thermocouple to be connected in turn.

Calibration over the temperature range expected was achieved by maintaining the junctions in the chamber at room temperature and varying the temperature of the water in the Dewar flask. Connections to the galvanometer, which had a

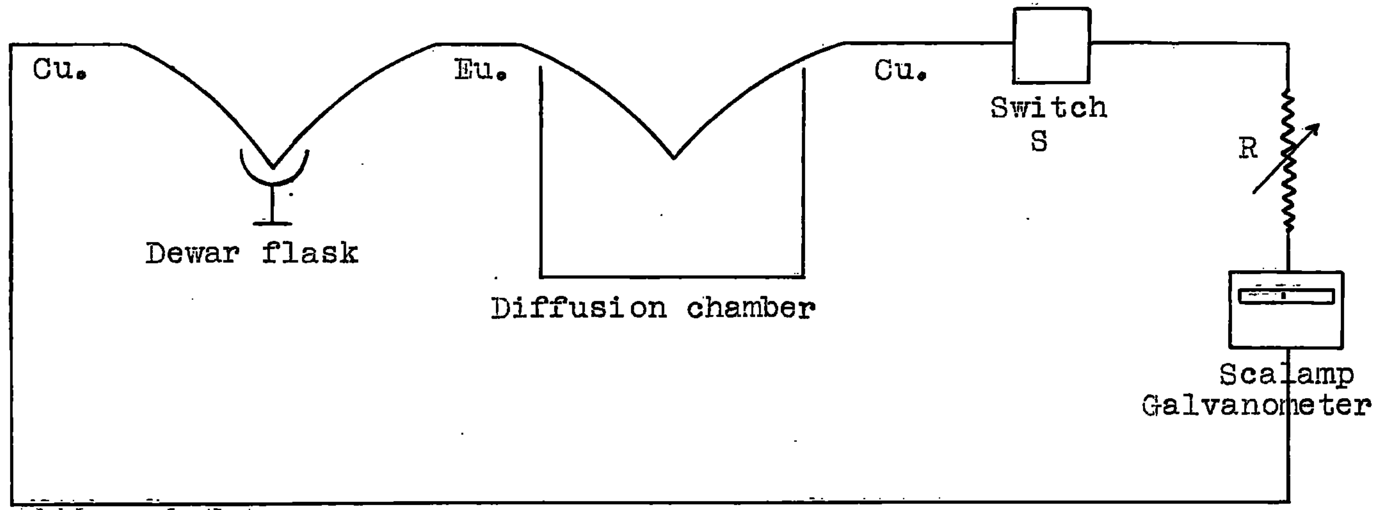


Fig. 4 Thermocouple Arrrangement

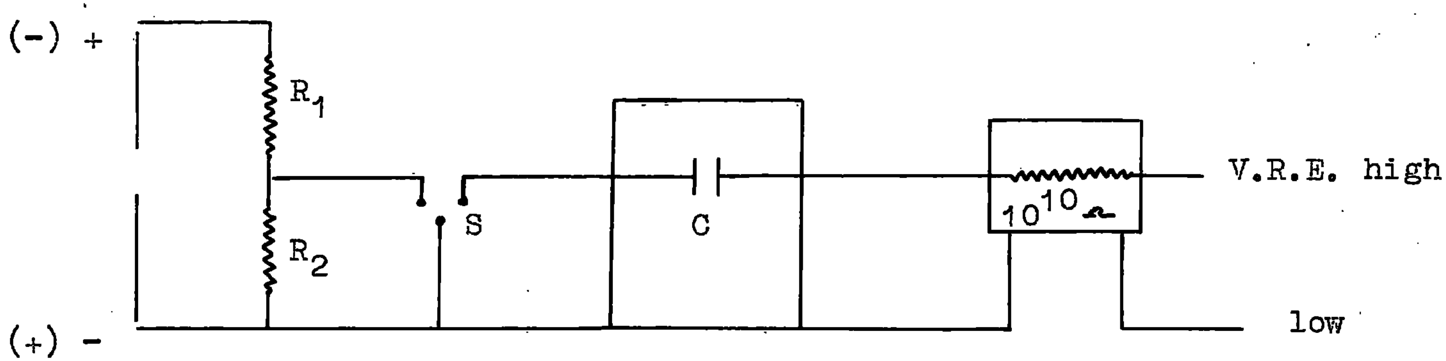


Fig. 5 V.R.E. Calibration

central zero scale, were made so that the deflection was negative when the chamber was colder than the Dewar flask. During the calibration a value of R was obtained which made a deflection of one division equivalent to a temperature difference of 1°C. Thus if a positive deflection, numerically equal to the room temperature, is set on the scale when there is no temperature difference the temperatures of the junctions can be read directly.

The temperature in the chamber increased quite steadily, typical values at 2 mm, 1½, 3, 4½, 6 and 7 cms. being -28, -20, -15, -11, -8 and -6°C. The graph in Fig. 3 shows that the increase is almost linear above the second thermocouple and thus the temperature at any level can be estimated. The lowest temperature obtained was -30°C.

The copper sheet (A in Fig. 2), which supports the water vapour source, is bolted to the top of the inner chamber making it essentially a separate unit. The water vapour source was made of crimped metal strips wound tightly together and fitted into a metal container. The crimping was achieved by forcing the metal between gear wheels with fine teeth, and its capillary action held sufficient water for three to four days work. A 33Ω 4W resistor run from a 12V a.c. supply acted as an efficient heater for the source and a thermocouple is fitted to measure its temperature.

The whole of the chamber is covered by a copper sheet

(B in Fig. 2), which is screwed onto a wooden platform built around the outer walls. A rubber seal was fitted between the platform and the copper sheet to prevent the influx of atmospheric nuclei, which would cause condensation and restrict the growth of the ice crystals. For the same reason all the gaps between the movable top (C in Fig. 2) and the outer walls were packed with foam rubber.

3. Measurement of charge

A vibrating-reed electrometer (Vibron 33B) is used to measure single charges by raising a charged particle into a Faraday cylinder. The induced charge leaks away through a 10^{10} ohm resistor connected across the terminals of the electrometer and produces a ballistic throw proportional to its magnitude. The 10^{10} ohm resistor is carefully screened in an earthed metal box. The Faraday cylinder is made of bronze and is suspended just above the top thermocouple in the inner chamber. The supporting wire runs along the axis of an earthed screening tube and is connected to the input terminal of the electrometer. The slightest movement of the coaxial cables used causes large deflections on the indicating voltmeter, but vibration of the motor was damped by the cork insulation and did not cause spurious fluctuations. The metal parts of the diffusion chamber provided sufficient electrostatic screening for the cylinder and the apparatus was quite stable electrically, random fluctuations being of the order of 0.1 mV on the 10 mV range.

The electrometer was calibrated by putting a condenser in place of the Faraday cylinder, applying a known voltage and discharging through the system. The apparatus used is shown in Fig. 5. The condenser was completely enclosed in an earthed metal box and the switch S, which was also screened had long flexible leads so that it could be depressed without producing any spurious charge. From Fig. 5 it can be seen that the charge on the condenser Q is $\frac{R_2}{R_1 + R_2}$ V.C., and as $R_1 + R_2$, V and C are constant Q is proportional to R_2. Fig. 6 shows the graphs of R_2 against deflection from which the sensitivity was calculated. The 10 mV and 30 mV ranges give 4.2×10^{-4} e.s.u. per mV and 4.0×10^{-4} e.s.u. per mV respectively. On the 10 mV range deflections of 0.1 mV can be read, but random fluctuations of this order occur and no deflection of less than 0.2 mV was accepted. This gives a limit of sensitivity of 8.4×10^{-5} e.s.u.

The electrometer was normally used on the 10 mV range set for positive input, but with the needle adjusted to read 5 mV so that positive and negative charges could be measured without adjustment. A positively charged particle put into the cylinder gives a positive deflection and a negative deflection when taken out. An average of these two deflections was always taken.

The disadvantage of the system is that if more than one

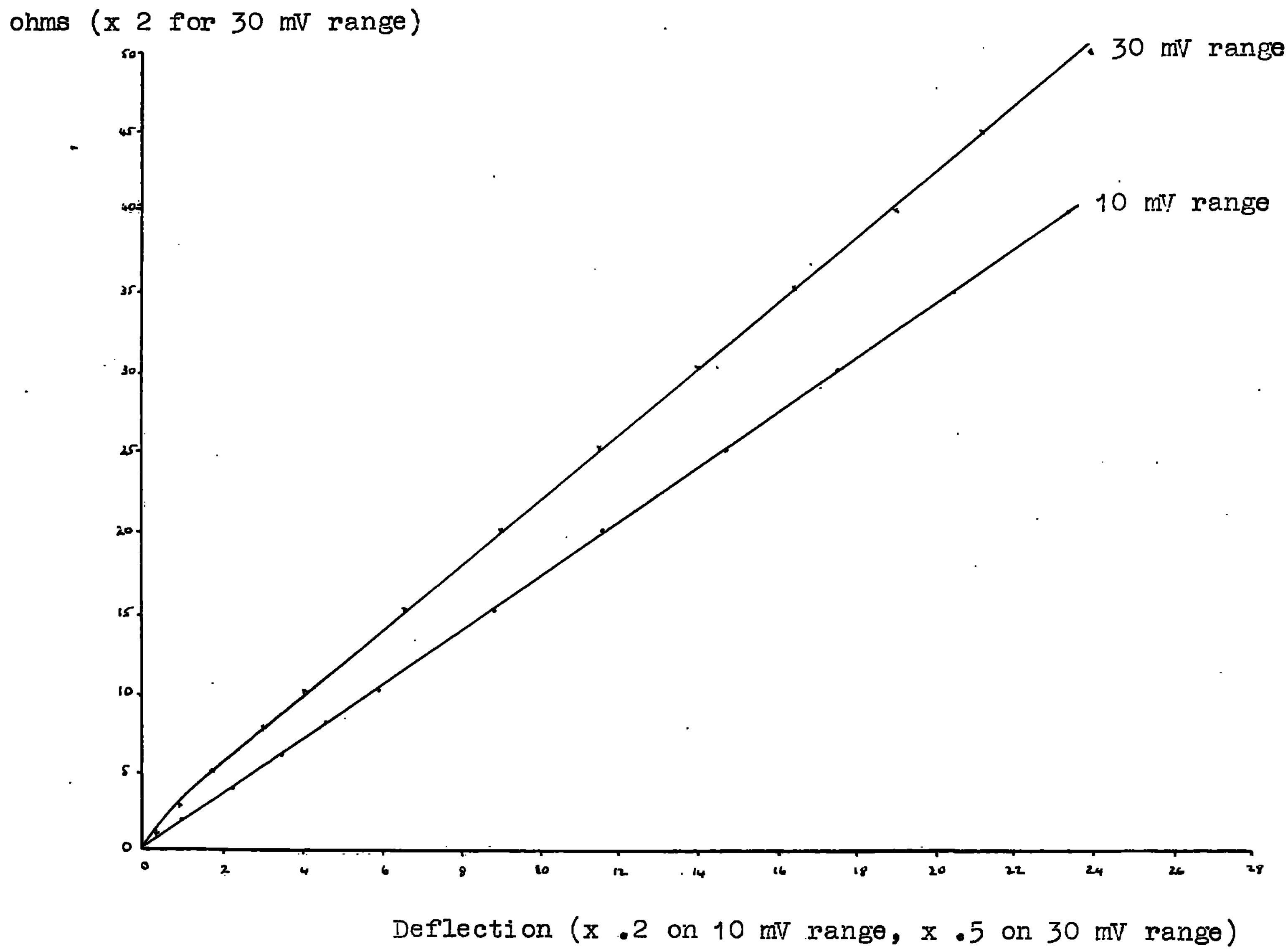


Fig. 6 V.R.E. Calibration Curves

charge is present on the fibre it is difficult to measure the magnitude of both. Fortunately the only time that more than one charged particle was observed on the fibre the charges were spurious.

CHAPTER FOUR

ICE-ICE CONTACT EXPERIMENTS

1. Introduction

Workman and Reynolds (1954) reported a charge separation of -5×10^{-4} e.s.u. per collision between an ice crystal and the riming spheres when there was a temperature difference of 2°C. Hutchinson (1960) used the apparatus described in Chapter three to investigate the charging produced by momentary contact of vapour grown ice crystals at different temperatures. The crystals were grown on an insulated fibre and a fine horizontal wire that could be rotated to make contact with the fibre. After contact between a suitable crystal on the fibre and those on the wire the fibre could be raised so that the crystal was inside the Faraday cylinder. Although temperature differences of 10°C were used no charging was detected. However, contact times of 0.2 to 0.5 seconds and sometimes as great as 2 seconds were reported, which are much greater than the time for maximum charge separation (0.01 seconds) deduced in the paper by Latham and Mason (1961a).

The work to be described here is a repetition of Hutchinson's experiments with general improvements in the apparatus, particularly an arrangement giving contact times of the order of 0.01 seconds. The main difficulties encountered with the apparatus were fogging and icing up, which seriously affected viewing and the slow growth of the ice crystals. The first was resolved by using the carefully sealed compartment described in Chapter Three, the second by fitting an improved water vapour source and taking precautions to prevent the influx of atmospheric nuclei.

2. <u>Contact mechanism</u>

The arrangement used is shown in Fig. 7. It consists of a rod supported in two machined guides and driven by a cam, which is rotated manually. The guides for the rod and the supports for the cam are bolted to a $\frac{1}{4}$" steel base to give rigidity. The device is bolted onto a metal platform and one of the guides, which fits flush with the outer wall of the diffusion chamber is screwed into it to prevent horizontal movement. Attached to the end of the rod is a stout wire, which passes through the perspex wall of the inner chamber and supports the fine horizontal wire on which the crystals were grown. The fine wire, which is at right angles to the supporting wire was situated approximately $\frac{1}{2}$" from the fibre. The throw of the cam is $\frac{1}{4}$" and the fine adjustment on the rod was used to move the wire forward so that contact was only just made.

The dimensions and construction of the cam are shown in Fig. 7. During the time that the point of contact, between

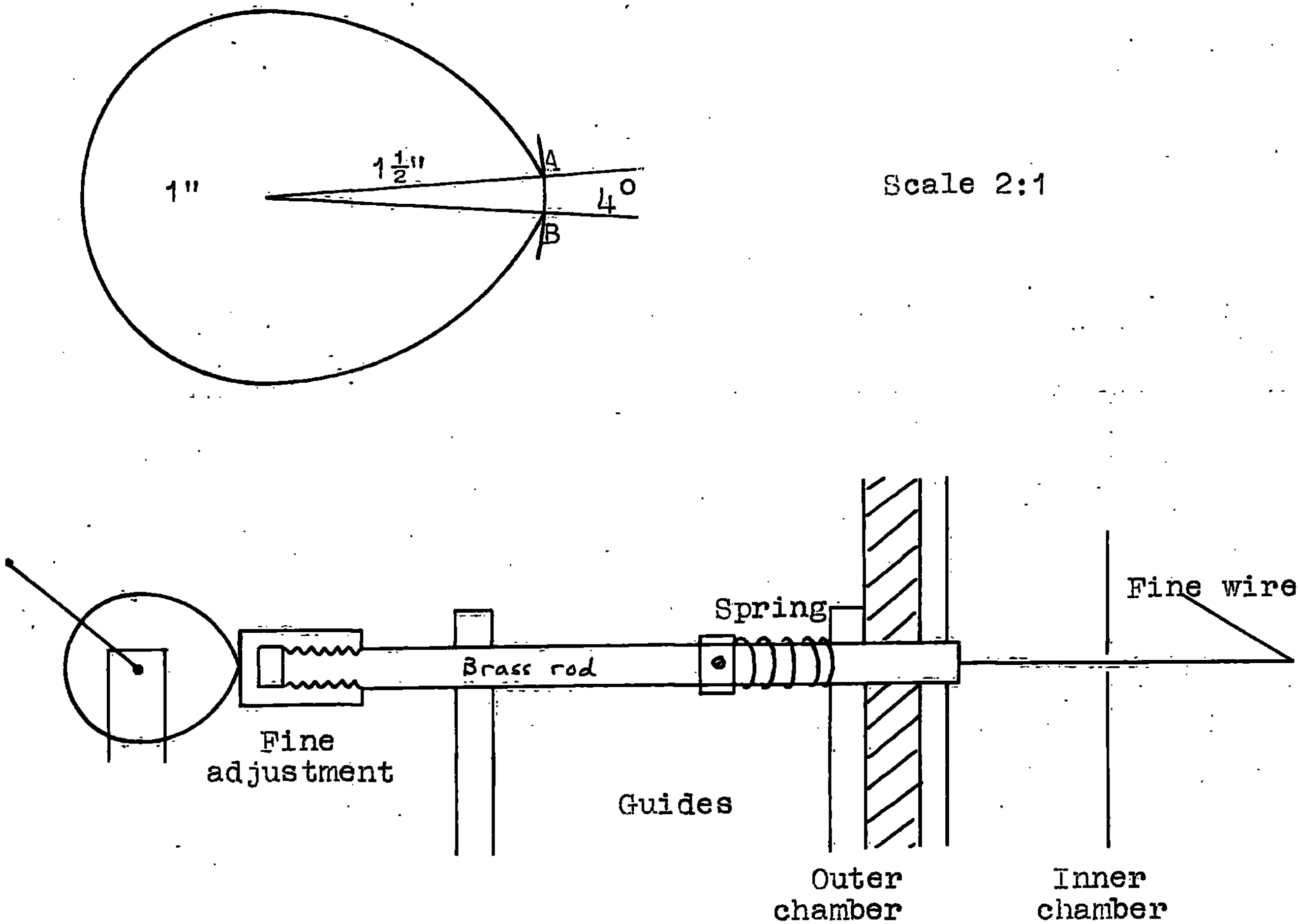


Fig. 7 Contact Mechanism

the cam and the rod, moves from A to B the rod will remain stationary. Thus if the time for one revolution is 1 second the rod will remain stationary for the order of 0.01 seconds. The rod is spring loaded so that it moves back quickly after contact has been made.

3. The fibre

Crystals were grown from the vapour on a fine insulated fibre attached to the brass rod shown in Fig. 2. The fibre was drawn out from a drop of Durofix adhesive placed on the rod and was held taut by a tiny solder weight. The rod could be raised or lowered so that any part of the fibre was inside the Faraday cylinder.

The properties of the fibre were carefully investigated. It was found that the fibre could be very roughly treated without becoming charged provided that it did not come into contact with the metal parts of the diffusion chamber. When this happened very persistent charges were produced and these were used to check that the charge-measuring arrangement was functioning satisfactorily. Any spurious charge could be removed from the fibre by bringing up a Cobalt 60 radioactive source which was available. The insulation of the fibre was very good, and with the rod and guide well lubricated with powdered graphite the fibre could be moved quickly without causing fluctuations of the electrometer pointer.

4. Crystal growth

It was found initially that no ice crystals grew at all on the fine wire of the contact mechanism due to heat flow along the supporting wire. A piece of polystyrene was then screwed onto the end of the rod and the supporting wire was attached to this, being earthed by the very fine fuse wire. When this was done crystals grew quite satisfactorily.

Crystals, on the fibre, grew to a length of a few millimetres in three to four hours after the introduction of the solid carbon dioxide into the paraffin tank. At this time the fine wire was covered with a 'layer' of ice, individual crystals not having grown to any appreciable size, and contact was duly made between a single crystal on the fibre and the layer of ice on the fine wire.

It may be mentioned here that the temperature dependence of crystal type, described by Hallett and Mason (1958), was clearly seen. Often the change from one type to another was observed when the position of the fibre had been altered, the most striking example being hexagonal plates growing on prismatic columns.

5. Experimental procedure

After the crystals had grown to a suitable size and a single crystal on the fibre had been chosen, the fibre was raised quickly through the Faraday cylinder to ensure that there was no initial charge on the ice. (No charging was ever observed due to the growth of the crystals alone). The crystal was then lowered and the contact mechanism adjusted so that contact could be made between the single crystal and the ice on the fine horizontal wire. The crystal was then lowered to its original position and left for a time to ensure that it was at the temperature of the surroundings.

The temperatures of the crystal and the ice on the wire were estimated from their positions in the chamber and noted down. The crystal was then raised quickly to the level of the wire and a single contact made. Immediately after contact the crystal was moved in andout of the Faraday cylinder and the electrometer observed for any deflection on the indicating voltmeter.

6. Results

Contact was made, both gently and violently, between crystals having temperature differences ranging from 0 to 10°C, but no charging was observed that could be associated with the contact. Small charges were observed when a large crystal was broken, but as this only occurred when contact was violent the charges were presumed to be spurious.

7. A balanced electrometer circuit

A preliminary investigation of the stability of a balanced electrometer tube circuit was carried out in order to test the possibility of building a D.C. amplifier with greater sensitivity than the electrometer arrangement. The circuit chosen is described by Dubridge (1931) and is essentially a Wheatstone's bridge arrangement with amplifier tubes in two arms. When correctly balanced the circuit can be made independent of battery fluctuations.

The circuit is shown in Fig. 8. A double electrometer valve (Ferranti DBM 4A) was used instead of the two tetrodes in the Dubridge circuit. As this valve has a common cathode and heater, no provision is made to cancel heater voltage variations. Grid bias batteries are used to supply grid and anode voltages and two well charged accumulators for the heater supply. High voltage wire wound resistors are used in the anode and grid circuits and a 10kΩ resistance box is used for the variable resistance R_1.

Initially the valve and grid resistors were carefully screened in a light tight metal box and a Scalamp galvanometer used to find the balance point. Balancing is achieved, with the circuit operating at normal voltages, in the following way. R_1 is adjusted to bring the galvanometer spot to zero and then a small change is made in the anode voltage. The deflection on the galvanometer is noted and then the value of R_1 changed by 1000Ω. The spot is brought back to zero by varying the various grid potentials and then the same change in the anode voltage made. If the deflection on the galvanometer is smaller then R_1 has been changed in the correct direction and the process is repeated until the change in anode voltage produces no deflection on the

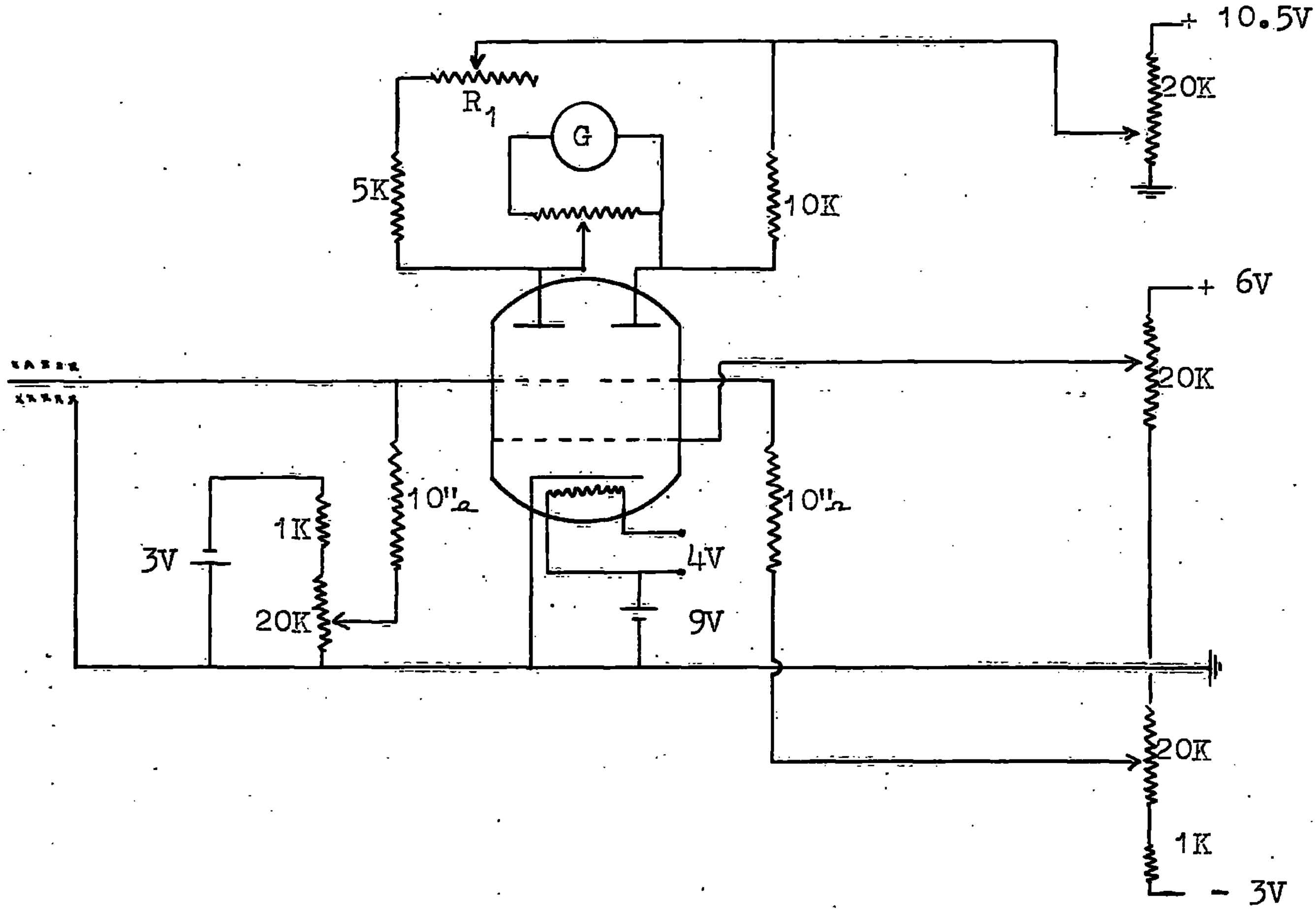


Fig. 8 Balanced Electrometer Tube Circuit

galvanometer. This balance point can be found quite quickly with practice and changes of $\frac{1}{2}$ volt on the anode give no deflection on the galvanometer.

The current sensitivity of the circuit $S_i = {}^d/_i$ where 'd' is the deflection on the galvanometer and 'i' the current in the grid resistor R. The change in the grid potential $\Delta e_g = R.i$ and the corresponding anode current change $\Delta ia = g_m \Delta e_g$. If 6 is the sensitivity of the galvanometer then the deflection $d = \Delta i_a\ 6$.

$$\text{Thus } S = \frac{d}{i} = \frac{\Delta i_a 6}{i} = \frac{g_m \Delta e_g\ 6}{i} = \frac{g_m\ R_1 i\ 6}{i}$$

$$\text{i.e. } \underline{S = g_m.R.6}$$

Using a Fluxmeter of sensitivity 1430 mm/μA, an input resistor of $10^{11}\Omega$ and knowing $g_m(60\mu A/_V)$ a theoretical current sensitivity of 8.58×10^{15} mm/A is obtained.

A preliminary calibration using the Scalamp galvanometer (6 = 67 mm/μA) was carried out and a charge of 10^{-13} coulombs gave a deflection of 4.5 mm, but random fluctuations were of the order of 1 mm.

In order to improve the screening a light tight metal box was constructed which housed the valve and grid resistors, batteries and anode resistors in separate compartments. Co-axial cables were used for all external connections and the only components outside the earthed box were the resistance box R_I, the accumulators and the galvanometer.

However, the stability was not noticeably increased and it was decided that the circuit would not readily give a sensitivity greater than that of the electrometer arrangement.

CHAPTER FIVE

THE NUCLEATION OF SUPERCOOLED DROPS

1. Introduction

Mason and Maybank (1960) investigated the freezing of supercooled water droplets and found that under certain conditions charge was separated. When drops were nucleated near $0^{\circ}C$ and frozen at a lower temperature a strong ice shell was formed and as freezing proceeded the pressure built up inside the drops often caused the drop to burst. It was found that charging always accompanied fragmentation of the drop. The explanation was that the radial temperature gradient across the shell caused a migration of protons from the warmer inner surface giving a charge separation across the shell. If an excess of the outer surface was carried away when the drop burst a negative charge would be left on the residue of the drop. The results showed that the residue charge was mainly negative, this being three times more frequent than positive, and an average charge of 10^{-3} e.s.u. per drop for 1 mm. diameter drops was obtained.

As referred to in Chapter 2, Latham and Mason (1961 b) followed their work on the charging of graupel due to impact of ice crystals by investigating the charging due to the accretion of supercooled droplets. The apparatus was constructed in a cold room, the temperature of which could be varied from 0 to - 17°C. Droplets of uniform diameter were drawn in an air stream past an ice-coated sphere, which was connected to a vibrating reed electrometer. The droplets fell several feet before reaching the sphere and their temperature reached that of the cold room. Both the temperature of the ice surface and the velocity of the air stream could be varied and the effects of these variables were investigated.

It was found that strong negative charging of the ice sphere resulted if the droplets were between 50 and 80 μ in diameter, but the charging decreased above and below these values. After low air stream velocities charging was observed reaching a maximum at 10 m./sec. After this the charging began to decrease due to positive charge produced by splashing and at 30 m./sec. positive charging was observed. For air temperatures in the range - 6 to - 17°C charging was constant but fell rapidly below - 6°C as the latent heat could not be dissipated and at air temperatures close to 0°C the ice sphere became wet.

2. Experimental Work

The experiment was intended to investigate the charging of a single drop nucleated by an ice sphere. The super-cooled drop was to be supported by a fibre and then brought into contact with a larger frozen drop on a second fibre.

After contact the two fibres could be raised through the Faraday cylinder so that any charging could be measured.

a) A spurious effect

The Durofix fibre is hydrophobic and in order to obtain drops of suitable diameter on the fibre ice crystals were grown from the vapour and then melted by raising above the $0^{\circ}C$ level. Suitable drops were obtained in this way, but it was found that charging had occurred. The procedure was repeated several times to ensure that the charging was due to the melting of the ice crystals. This was found to be so, but although two distinct charges, negative above positive on the fibre were observed it was difficult to determine their exact position. A typical example of the fibre after melting was, 1 cm. of unmelted ice crystals, above this 1 cm. of wet ice and droplets with an associated charge of $4 \cdot 6 \times 10^{-3}$ e.s.u. a gap of $1\frac{1}{2}$ cms. and then a few small droplets with a charge of $3 \cdot 4 \times 10^{-3}$ e.s.u. apparently associated with them.

The production of these charges seemed to be connected with the work reported by Dinger and Gunn (1946) who found that when an ice mass is melted it gains a positive charge, an equal negative charge going into the air. Their explanation was that bubbles of air trapped during the freezing became negatively charged as they passed through the melted ice and carried this charge into the air, leaving the water with a resultant positive charge.

Further experiments were carried out to try to relate the magnitude of the charge with the quantity of ice melted. It was found that if the ice melted to give small droplets equally spaced on the fibre no charging was observed, but if the droplets coalesced a large quantity of charge was separated. It seemed from this that the charging was due to the movement of the droplets over the insulating fibre rather than the melting of the ice.

In order to test this a fibre was constructed with constrictions near the top so that a drop could be placed on the fibre and then moved to a position where the slightest movement caused the drop to run down the fibre. Any charge on the drop was removed with the Cobalt 60 source. Using this fibre and drops at room temperature it was found that a charge separation occurred similar to that found in the 'melting ice' experiments. Drops at $0^{\circ}C$ also gave a similar separation. Over sixty drops were investigated and the results will be summarised.

Charge was always produced with positive charge on the drop (maximum value 3×10^{2} e.s.u. for a 1 mm. diameter drop moving 4 cms. down the fibre) and a negative charge of approximately the same value on the fibre. There was definite evidence that the negative charge occurred in a limited region around the initial position of the drop, but although the fibre was examined closely there was no evidence of any smaller droplets left behind. No relation could be found between the charge separated and the distance moved down the fibre or the rate of fall.

These results suggest that the charging observed when the ice crystals were melted was due to the movement of the resulting droplets and was not connected with the change of state.

b) <u>Experimental procedure</u>

Drops were suspended from a thicker Durofix fibre attached to the bottom of the weight which kept the insulating fibre taut. A second guide and supporting rod was soldered to the one used in the ice-ice contact experiment and was used to support the frozen drop. This was suspended in the same way from a fibre attached to a stouter wire fixed at right angles to the supporting rod. Careful positioning of the fibres allowed the drops to be brought into contact when the second supporting rod was rotated.

In an actual experiment the water drop was held just below the $0^{\circ}C$ level and the frozen drop was allowed to reach a temperature of around $-15^{\circ}C$. The supercooled drop was then lowered quickly to the level of the frozen drop, contact made and the two were raised into the Faraday cylinder when freezing was completed. No charging at all was observed and the water drop became opaque immediately contact was made. The drop showed no sign of spicule production. The water drop also spread on the frozen drop and it was thought that this was because the small heat capacity of the frozen drop could not dissipate the latent heat liberated. A large ice mass was produced by freezing a large water drop held in a fine wire loop and contact was made by lowering the supercooled drop on to it. However it was found that spreading still occurred and again the drop became opaque almost immediately. The diamater of the drop was decreased from 1 mm. to 250μ, but the same type of freezing occurred.

It seemed that the ice mass was acting as a large heat sink and the large area of contact allowed the latent heat to be dissipated very quickly. This increase in the rate of freezing would result in an initial ice shell not suitable for the formation of spicules and fragmentation (Mason and Maybank 1960). The rates of freezing of drops in free air and in contact with an ice mass were compared by supporting millimetre drops from one junction of a fine thermocouple.

The results showed that the rate of freezing was increased by a factor of 10 and this would seriously deter the formation of spicules.

It was realised that very small droplets might remain spherical and thus reduce the area through which the latent heat could be dissipated. A spray was constructed which consisted of a jet of air being blown against a jet of water, both produced from a test tube fitted with hand bellows.

The spray did not give drops of uniform diameter, but this proved useful as drops of different diameter could be compared.

An ice mass held in a wire loop was fixed at some level in the diffusion chamber and was allowed to reach the temperature of the environment. The spray was fitted with an ice-water mixture and kept in an ice-water bath to keep the temperature as near $0^{o}C$ as possible. A small cover was removed from the top of the chamber and the ice mass was placed directly below. Drops were then sprayed above the opening and allowed to fall slowly onto the ice mass. The drops were viewed through the microscope after freezing and the appearance and size noted down.

3. Results

It was found that the smaller droplets remained spherical after freezing and were transparent. Larger droplets were opaque and had spread on the surface of the ice mass, but appeared pointed at the surface away from the ice mass. Although there was some overlap between the two classes all droplets below 66 μ diameter (1 division on the microscope eyepiece graticule) were spherical and transparent and all above 130 μ were opaque.

The temperature of the ice mass was varied and at $-9^{o}C$ the number of spherical drops began to decrease and at $-6^{o}C$ no spherical droplets could be seen at all.

An attempt was made to remove the charge, due to spraying, on the droplets by applying a voltage to the water in the spray, but although the sign could be reversed it was not possible to remove all the charge.

CHAPTER SIX

FRAGMENTATION AND ELECTRIFICATION OF SUPERCOOLED DROPS

In the paper described in Chapter 5 Mason and Maybank (1960) reported that as the drop diameter decreased from 1 mm. to ·35 mm. the charge produced fell by a factor of two, but the data were insufficient to determine the dependence of charge on size. The present apparatus could easily be adapted to repeat Mason and Maybank's work and it was hoped that a relationship between drop diameter and the magnitude of the charge produced could be found.

1. Experimental technique

It was necessary that the drops be cooled to just below $0^{o}C$ and then nucleated, at the same temperature, in an environment well below $0^{o}C$. The drops were supported by a Durofix fibre as in the previous experiment and could, therefore, be moved quickly to any level in the diffusion chamber. A length of glass tubing was fixed in the second guide, the lower end being level with the bottom of the Faraday cylinder. A small piece of solid carbon dioxide dropped down this glass tube produced a cloud of ice crystals below the Faraday cylinder so that the supercooled drop was nucleated as it was lowered into position.

The fibre supporting the drop was constructed so that the drop hung freely from the end, i.e. the fibre did not penetrate far into the drop. This was achieved by attaching a small blob of Durofix to the end of the fibre and cutting it to the shape of a squat pyramid. This was found to be necessary as the drop shattered into two pieces and on occasions the fibre, frozen into both, prevented complete separation.

The drops were nucleated around $-2^{\circ}C$, and in every experiment the environment temperature was $-15^{\circ}C$. Any charge produced was measured by moving the drop into and then out of the Faraday cylinder and taking an average of the two deflections. Care was taken to ensure that the drops were not charged before nucleation, any charge present being removed with the Cobalt 60 source.

2. Observations

a) Freezing process

Immediately after the drop was nucleated a thin layer of transparent ice was formed on the surface. This grew slowly and on occasions a leading edge of ice could be seen to move over the drop. Usually this film was smooth with only the occasional irregularity, but at times there were numerous ridges presumably because the film had been formed more quickly. Cracks then appeared in the ice film and water could be seen to spread and freeze on the surface of the drop. In this way the ice film thickened, becoming slightly less transparent, until a point was reached when the ice film could not crack easily. A bulge then formed, usually around

Fig. 9a

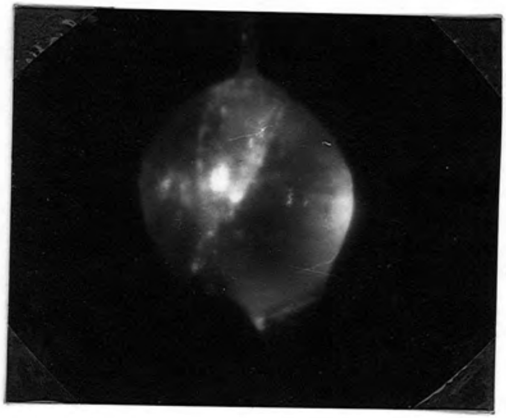


Fig. 9b

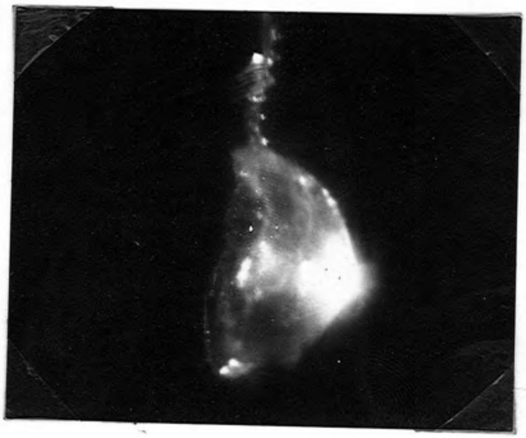


Fig. 9c

a crack or ridge on the ice surface, and was more opaque than the rest of the drop. The bulge continued to grow slowly until a spicule appeared which grew comparatively quickly. The spicule was always transparent and grew sometimes steadily, but more often in a series of sudden steps. Occasionally, towards the end of the growth of the spicule, water was seen to run down the outside and freeze around the base of the spicule. Fig. 9a shows a typical spicule. The base can be seen to be much more opaque than the rest of the drop.

After the growth of the spicule freezing proceeded much more slowly and continued with the sudden appearance of large cracks in the ice shell. Again water could be seen to spread and freeze on the surface of the drop and also numerous bubbles were liberated inside the drop and the spicule. The freezing continued in this way and the final freezing was often accompanied by a large crack completely encircling the drop. An example of this is shown in Fig. 9b ; in this case the final crack forced two halves of the drop apart.

b) <u>Fragmentation</u>

Two types of fragmentation were observed. In one the drop was split into two parts by a large crack in the final stages of freezing. Fig. 9c shows the remains of a drop which had shattered in this way. The opaque ice in the centre of the drop was probably formed by the water present in the drop when fragmentation occurred. In the second type observed the spicule was ejected, usually broken at right angles to its axis. For both types fragmentation was violent causing the fibre to twist violently.

c) Charge production

In all but one case charge was detected when fragmentation of the drop occurred. No charge was detected if fragmentation did not occur so that the freezing process and impact of excess ice crystals could not be the cause of the charging. On a number of occasions the drop was split open by a large crack and the same violent twisting of the fibre occurred, but only once was a charge detected on the drop. This rules out the possibility of the charge being due to the movement of the fibre.

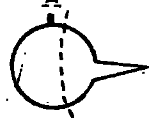
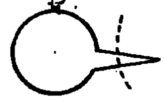
3. Results

Table 1 shows the charges produced by the two types of fragmentation observed and these will be discussed separately.

Column A shows the charging observed when splitting of the drop took place. In all but two cases marked with an asterisk the spicule was carried away on the part of the drop ejected. With the exception of the value underlined all the charges are for the major residue type described by Mason and Maybank (1960). The results show that the charging for this type is mainly negative, the negatively

TABLE 1

Drop diameter 1 - 1.5 mm.

	A		B		C
Type of Shattering					
Charge on residue	10^{-3} e.s.u.		10^{-3} e.s.u.		10^{-3} e.s.u.
	- 25	+ 2.0*	- 3.7	+ 11.3	- .3
	- 8.4	+ .55	- 3.4	+ 5.0	X
	- 2.1	+ .42	- 2.7	+ 1.2	
	- 1.9	+ .3*	- 2.1	+ 1.2	
	- 1.4		- 1.7	+ .6	
	- .7		- 1.5	+ .4	
	- .6		- 1.3	+ .3	
	- .4		- 1.3	+ .3	
	- .4		- 1.3	+ .2	
	- .2		- .9	+ .1	
	0		- .9	+ .1	
			- .9		
			- .8		
			- .4		
			- .4		
			- .2		
			- .1		
			- .1		

* on five other occasions no charging was observed

charged drops being almost three times more frequent. The average value of the negative charge per drop is $1 \cdot 8 \times 10^{-3}$ e.s.u. and the average value of the positive charge per drop is $0 \cdot 82 \times 10^{-3}$ e.s.u.

Column B shows the charging observed when the spicule was broken and a part ejected. In this type positive charging is more frequent, but negative charging appears to be stronger. The average value of the negative charge per drop is $1 \cdot 3 \times 10^{-3}$ e.s.u. and the average positive charge per drop is $0 \cdot 9 \times 10^{-3}$ e.s.u.

Column C has been included because although the two parts of the drop were not fully separated some splintering would be expected.

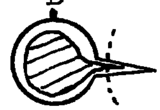
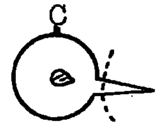
Table 2 shows the results obtained with larger drops placed nearer the front of the diffusion chamber so that the mechanism of the freezing process could be observed more clearly.

Column A shows the charging observed when splitting of the drop took place. No positive charging was observed with these larger drops.

Column B shows the charging observed when the spicule was broken and ejected almost immediately after being formed and before any further freezing of the drop took place. The values marked with an asterisk are for cases in which a small amount of the freezing took place before the spicule was ejected.

<u>TABLE 2</u>

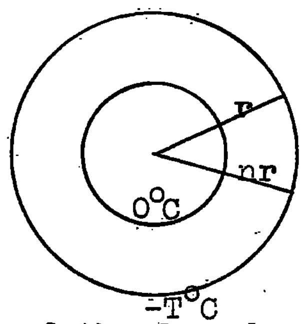
<u>Drop diameter</u> 1.3 - 1.5 mm.

	A	B		C
Type of Shattering				
Charge on residue	10^{-3} e.s.u.	10^{-3} e.s.u.		10^{-3} e.s.u.
	- 25	- 2.1	+ .42*	- .4*
	- 2.1	- 1.3	+ .13*	+ 11.3
	- 1.9	- 1.3		+ 5.0
	- 1.4	- 1.3		+ .6
	- .6	- .9		+ 1.2*
	- .4	- .9		+ .3*
		- .4		
		- 2.7*		
		- 1.5*		
		- .17*		

Column C shows the charging observed when the spicule was broken and ejected with the final freezing of the drop. This type was often associated with a large crack which passed through the spicule and the drop. The values marked with an asterisk are for cases in which a small amount of the freezing took place after the spicule had been ejected.

4. Discussion of the results

It is interesting to compare the results listed in Table 1 with the total charge separation in the ice shell predicted by Latham and Mason's theory.



Let the radius of the drop be r and let the thickness of the ice shell be nr so that n is the fraction of the radius in the ice. Assuming a uniform thickness of the ice shell and a uniform temperature gradient across it, Latham and Mason's theory predicts a charge separation of $q = 4{\cdot}9 \times 10^{-5} \frac{\Delta T}{nr}$ e.s.u./cm.^{2} where ΔT is the temperature difference between the inner and outer surfaces of the ice shell.

If the area through which the charge is being separated is taken to be equal to the surface area of a sphere of radius $(r - \frac{nr}{2})$ then the total charge separated is given by $4{\cdot}9 \times 10^{-5} \frac{\Delta T}{nr} 4\pi(r - \frac{nr}{2})^2$ e.s.u. Simplifying using a value of $\Delta T = 15^{\circ}C$ we have $Q = 9{\cdot}3 \times 10^{-3}\, r(\frac{n}{4} + \frac{1}{n} - 1)$ e.s.u. where Q is the total charge separated across the ice shell. The average radius in Table 1 is 0·63 mm. and Fig. 9c gives $n = \frac{3}{4}$. Substituting these figures a value for Q of $0{\cdot}3 \times 10^{-3}$ e.s.u. is obtained.

This suggests that the maximum charging possible, assuming <u>all</u> the charge of one sign to be ejected, is $0{\cdot}3 \times 10^{-3}$ e.s.u. However, in 86% of the cases the observed charge was greater than this and in 16% of the cases the observed charge was greater by more than a factor of 10.

The ways in which the predicted charge could be increased will be considered.

(1) <u>Increase in the temperature gradient</u>

a) <u>Temperature of the outer surface of the ice shell</u>

This could be estimated from the position of the drop in the diffusion chamber to within $\pm 1^{\circ}C$ and no systematic error could have occurred in estimating the drop's position. The drop may have lost heat to the cold copper base by radiation, but this would be cancelled out by radiation received from the warmer copper cover of the chamber.

b) <u>The temperature of the inner surface of the ice shell</u>

It seems improbable that the water could be warmer than $0^{\circ}C$, and the effects of pressure and impurities in the water would tend to cause supercooling and therefore decrease the temperature gradient.

c) Thickness of the ice shell

When the drop was split, fragmentation always occurred after the majority of the freezing had taken place so that the drop in Fig. 9c will be fairly typical. However, the effect of a decrease in the ice shell thickness can be calculated from the equation $Q = 9{\cdot}3 \times 10^{-3}\ r\left(\frac{n}{4} + \frac{1}{n} - 1\right)$ e.s.u.

n	Q e.s.u.	%
$\frac{3}{4}$	0.3×10^{-3}	20
$\frac{1}{2}$	0.66×10^{-3}	53
$\frac{1}{3}$	1.2×10^{-3}	60
$\frac{1}{4}$	2.4×10^{-3}	86

% – percentage of the observed values of charging in Column A accounted for by the predicted value.

If n is decreased further all the observed charges can be accounted for, but it must be stressed that the values quoted above are for the TOTAL charge separated in the ice shell and only a part of this will be removed when the drop bursts.

It seems unlikely that an increase in the temperature gradient alone could account for the difference between the observed and the predicted charging.

(2) The effects of impurities

Mason and Maybank (1960) suggested that the increase in conductivity caused by the presence of impurities in the ice could account for the high values of charging found in their work. In a later paper Latham and Mason (1961 a) described the results of an experiment to determine the effects of impurities on the charge separation. It was reported that hydrogen flouride and carbon dioxide gave an increase in the charge separation whereas sodium chloride caused a decrease. The maximum increase was caused by hydrogen fluoride, a 10^{-3} Molar solution giving an increase of 55% for a temperature difference of 6^{o}C. For the same temperature difference ice saturated with carbon dioxide gave an increase of 13% on the theoretical charging.

The presence of impurities in the ice would not, therefore, account for the high values of charging observed.

(3) Frictional effects

Mason and Maybank (1960) also suggested that friction between the ice surfaces might play an important part in the charging mechanism. If this were so a marked contrast between the magnitudes of the charges in Column A and B would be expected because of the difference in area of contact involved. However, no such contrast is evident.

(4) The effect of the spicule

In the early stages of growth a large temperature gradient will be built up across the ice shell of the spicule and this may produce sufficient charge separation to account for the values in Column B. Using the dimensions of the spicule in Fig. , and assuming a temperature difference of 15°C between the inner and outer surfaces, the thickness of the shell required for fixed charge separation can be determined from the equation $q = 4{\cdot}95 \times 10^{-5} \frac{dt}{dx}$ e.s.u./cm.^{2}

Charge separated	Thickness of shell
10^{-3} e.s.u.	25 μ
2×10^{-3} e.s.u.	12·5 μ
3×10^{-3} e.s.u.	8·3 μ

Unless the spicule is ejected immediately it is formed it seems unlikely that sufficient charge could be separated to account for the observed values. Also only part of this charge will be removed when the spicule is broken and ejected.

None of the effects considered above can account for the discrepancy between the predicted and the observed charging.

The results in Table 2 suggest that the sign of the charge carried away when the spicule is ejected is determined by the amount of freezing that has taken place. In Column B the spicule was still transparant when fragmentation took place and must have contained a large quantity of liquid water. In Column C the majority of the freezing had taken place so that the spicule must have contained little or no liquid water. The obvious difference of the signs in Columns A and B suggests that the water may play an importance role in the charge-separating mechanism inside the drop.

Workman and Reynolds (1954) reported that water containing the impurities found in natural hail in New Mexico gave a charge separation of 10^4 e.s.u. per c.c. of water frozen with positive water and negative ice. Using this value the charge separated in a 1 mm. diameter drop when the thickness of the ice shell is ·25 mm. can be calculated

$$\text{Volume of water frozen} = \frac{4}{3}\,\bar{n}(5^3 - 2{\cdot}5^3)\ 10^{-6}\ \text{c.c}$$

$$= 4{\cdot}6 \times 10^{-4}\ \text{c.c.}$$

∴ Charge separated is 4·6 e.s.u.

Mason (1956) reported that when a drop is nucleated in oil a fine jet of water can be seen to be ejected when the outer shell bursts. If the Workman-Reynolds effect was in operation this jet of water would carry away positive charge leaving the drop with a resultant negative charge. Negative charge could be carried away by ice ejected by the sudden freezing of a quantity of water which remained on the drop. This would leave the drop positively charged.

This suggested mechanism can account for all the observed effects both qualitatively and quantitatively if the Workman and Reynolds value of 10^4 e.s.u. per c.c. frozen holds.

(5) Conclusions

When a supercooled drop is nucleated near 0°C and frozen at a lower temperature fragmentation accompanied by charging of the drop occurs. The charging appears to be too large to be accounted for by Mason and Maybank's theory, values an order of magnitude greater than the predicted charge separation over the whole drop having been measured. Factors which may increase the theoretical value cannot account for the difference.

It appears that when part of the spicule is ejected the amount of water present determines the sign of the charging. This suggests that the charge separating mechanism inside the drop involves the liquid water present. The effect reported by Workman and Reynolds (1950) could account for the amounts of charge observed.

ACKNOWLEDGEMENTS

The writer wishes to express his gratitude to Dr. W.C.A. Hutchinson, and Dr. J.A. Chalmers for their guidance and encouragement, and to Professor G.D. Rochester for the research facilities made available in the Durham Colleges.

He thanks the laboratory technicians for their ready assistance and particularly Mr. Owen of the Chemistry Department, who arranged the supply of solid carbon dioxide. Special thanks are due to the friends and fellow research workers who made many valuable suggestions during discussions of the work.

The writer is indebted to D.S.I.R. for the Advanced Studentship held during the period October, 1959 to September, 1961.

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